

FORM PTO-1390
(REV 10-94)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

5848.170USWO

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

unknown 10/018644

INTERNATIONAL APPLICATION NO.

PCT/SE00/01334

INTERNATIONAL FILING DATE

22 June 2000

PRIORITY DATE CLAIMED

1 July 1999

TITLE OF INVENTION

AN INSULATION COMPOSITION FOR AN ELECTRIC CABLE

APPLICANT(S) FOR DO/EO/US

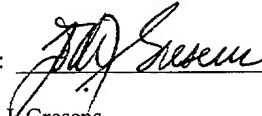
GUSTAFSSON et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(l).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An unsigned oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information: International Publication Page, Form PCT/RO/101, Form PCT/ISA/210, Form PCT/IB/308, Form PCT/IPEA/409 with amended sheets 24-25, 2 sheets of Drawing Pages, Preliminary Amendment, Marked-up Copy, Abstract

U.S. APPLICATION NO (If known, see 37 CFR 1.5) unknown 10/018644		INTERNATIONAL APPLICATION NO PCT/SE00/01334		ATTORNEY'S DOCKET NUMBER 5848.170USWO	
17. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a) (1)-(5)): Search Report has been prepared by the EPO or JPO.....\$890.00 International preliminary examination fee paid to USPTO (37 CFR 1.492(a)(1)).....\$710.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....\$740.00 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(3)) paid to USPTO \$1040.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)\$100.00				CALCULATIONS PTO USE ONLY	
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$1040.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	9 -20 = 0		X \$18.00	\$0.00	
Independent claims	2 -3 = 0		X \$84.00	\$0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$260.00	\$	
TOTAL OF ABOVE CALCULATIONS =				\$1040.00	
Reduction by 1/2 for filing by small entity, if applicable. Small entity status is claimed pursuant to 37 CFR 1.27				\$	
SUBTOTAL =				\$1040.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				+	\$
TOTAL NATIONAL FEE =				\$1040.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+	\$
TOTAL FEES ENCLOSED =				\$1040.00	
				Amount to be: refunded	\$
				charged	\$
a. <input checked="" type="checkbox"/> Check(s) in the amount of \$1040.00 to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-2725.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO John J. Gresens MERCHANT & GOULD P.O. Box 2903 Minneapolis, MN 55402-0903					
				SIGNATURE:  NAME: John J. Gresens	
REGISTRATION NUMBER: 33,112					

S/N unknown

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: GUSTAFSSON et al. Serial No.: unknown
Filed: 18 December 2001 Docket No.: 5848.170USWO
Title: AN INSULATION COMPOSITION FOR AN ELECTROC POWER CABLE

CERTIFICATE UNDER 37 CFR 1.10

'Express Mail' mailing label number: EV037638330US

Date of Deposit: 18 December 2001

I hereby certify that this correspondence is being deposited with the United States Postal Service 'Express Mail Post Office To Addressee' service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.

By: 

Name: Chris Stordahl

PRELIMINARY AMENDMENT

Box PCT
Assistant Commissioner for Patents
Washington, D. C. 20231

Dear Sir:

In connection with the above-identified application filed herewith, please enter the following preliminary amendment, based on claims amended in prosecution of the international application and published in the International Preliminary Examination Report, a copy of which is enclosed herewith:

IN THE ABSTRACT

Insert the attached Abstract page into the application as the last page thereof.

IN THE SPECIFICATION

A courtesy copy of the present specification is enclosed herewith. However, the World Intellectual Property Office (WIPO) copy should be relied upon if it is already in the U.S. Patent Office.

IN THE CLAIMS

Please amend claims 3 and 5-8 as follows:

3. (Amended) An insulating composition as claimed in claim 1, wherein the multimodal ethylene copolymer has a viscosity of
2500-7500 Pa.s at 135°C and a shear rate of 10 s⁻¹,
1000-2200 Pa.s at 135°C and a shear rate of 100 s⁻¹, and
250-400 Pa.s at 135°C and a shear rate of 1000 s⁻¹.
5. (Amended) An insulating composition as claimed in claim 1, wherein the comonomer of the copolymer is at least one member selected from the group consisting of propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, and 1-octene.
6. (Amended) An insulating composition as claimed in claim 1, wherein the MWD is 4-5.
7. (Amended) An insulating composition as claimed in claim 1, wherein the multimodal ethylene copolymer is a bimodal ethylene copolymer comprising 30-60 % by weight of a low molecular weight ethylene copolymer fraction and 70-40 % by weight of a high molecular weight ethylene copolymer fraction.
8. (Amended) An insulating composition as claimed in claim 1, wherein the multimodal ethylene copolymer includes a low molecular weight ethylene copolymer fraction having a density of 0.900-0.950 g/cm³ and a MFR₂ of 50-100 g/10 min.

REMARKS

The above preliminary amendment is made to remove multiple dependencies from claims 3 and 5-8.

A new abstract page is supplied to conform to that appearing on the publication page of the WIPO application, but the new Abstract is typed on a separate page as required by U.S. practice.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page is captioned "Marked-up Copy".

Applicants respectfully request that the preliminary amendment described herein be entered into the record prior to calculation of the filing fee and prior to examination and consideration of the above-identified application.

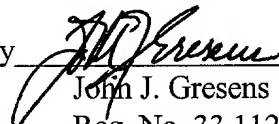
If a telephone conference would be helpful in resolving any issues concerning this communication, please contact Applicants' primary attorney-of record, John J. Gresens (Reg. No. 33,112), at (612) 371.5265.

Respectfully submitted,

MERCHANT & GOULD P.C.
P.O. Box 2903
Minneapolis, Minnesota 55402-0903
(612) 332-5300

Dated: 18 December 2001

By


John J. Gresens
Reg. No. 33,112

JJG:hjh

MARKED-UP COPY

3. An insulating composition as claimed in [any one of claims 1-2,] claim 1, wherein the multimodal ethylene copolymer has a viscosity of 2500-7500 Pa.s at 135°C and a shear rate of 10 s^{-1} , 1000-2200 Pa.s at 135°C and a shear rate of 100 s^{-1} , and 250-400 Pa.s at 135°C and a shear rate of 1000 s^{-1} .

5. An insulating composition as claimed in [any one of claims 1-4,] claim 1, wherein the comonomer of the copolymer is at least one member selected from the group consisting of propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, and 1-octene.

6. An insulating composition as claimed in [any one of claims 1-5,] claim 1, wherein the MWD is 4-5.

7. An insulating composition as claimed in [any one of claims 1-6,] claim 1, wherein the multimodal ethylene copolymer is a bimodal ethylene copolymer comprising 30-60 % by weight of a low molecular weight ethylene copolymer fraction and 70-40 % by weight of a high molecular weight ethylene copolymer fraction.

8. An insulating composition as claimed in [any one of claims 1-7,] claim 1, wherein the multimodal ethylene copolymer includes a low molecular weight ethylene copolymer fraction having a density of $0.900\text{-}0.950 \text{ g/cm}^3$ and a MFR₂ of 50-100 g/10 min.

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AN INSULATION COMPOSITION FOR AN ELECTRIC POWER CABLEField of the invention

The present invention relates to an insulating composition for an electric power cable which comprises a crosslinkable ethylene polymer. The present invention also relates to an electric power cable comprising a conductor surrounded by an inner semiconducting layer, an insulating layer, and an outer semiconducting layer

Background of the invention

Electric power cables for medium voltages (6-69 kV) and high voltages (> 69 kV) normally include one or more metal conductors surrounded by an insulating material like a polymer material, such as an ethylene polymer. In power cables the electric conductor is usually coated first with an inner semiconducting layer followed by an insulating layer, then an outer semiconducting layer followed by water barrier layers, if any, and on the outside a sheath layer. The layers of the cable are based on different types of ethylene polymers which usually are crosslinked.

A power cable of the above type is normally produced in the following way:

Three layers, one inner semiconductive layer, one insulating layer and one outer semiconducting layer, are extruded onto a conductor using a triple head extruder. In this construction the insulation layer is imbedded inbetween the semiconductive layers like a sandwich construction. The insulation layer itself is normally one single layer. The thickness of the different layers depend on the gradient and the rating that the cable is exposed to. Typical values for the thickness of a MV/HV (medium and high voltage) construction are the following: the semiconductive layers are about 0.5-2 mm each and the insulation layer about 3-30 mm.

The three layers are normally extruded onto the conductor at a low temperature (below 135°C) in order to prevent the crosslinking reactions from taking place during the extrusion process. After the extrusion step
5 the construction is crosslinked in a pressurised vulcanising tube at an elevated temperature.

LDPE (low density polyethylene), i.e. polyethylene prepared by radical polymerisation at a high pressure and crosslinked by adding a peroxide in connection with the
10 extrusion of the cable, is today the predominant cable insulating material. Radical polymerisation results in long chain branched polymers having a relatively broad molecular weight distribution (MWD). This in turn results in desirable rheological properties with regard to their
15 application as insulating materials for electric power cables.

A limitation with LDPE lies in the fact that it is made by radical polymerisation. Radical polymerisation of ethylene is carried out at high temperatures of up to
20 about 300°C and at high pressures of about 100-300 MPa. To generate the high pressures needed energy consuming compressors are required. Considerable investment costs are also required for the polymerisation apparatus which must be able to resist the high pressures and tempera-
25 tures of radical initiated high pressure polymerisation.

With regard to insulating compositions for electric power cables it would be desirable both from a technical and an economical point of view if it was possible to make an ethylene polymer with the advantageous properties
30 of LDPE, but which was not made by radical polymerisation. This would mean that insulation for electric cables could be made not only at plants for high pressure polymerisation of ethylene, but also at the many existing plants for low pressure polymerisation of ethylene. In
35 order to be a satisfactory replacement for LDPE such a low pressure material would have to fulfil a number of requirements for insulating materials, such as good pro-

cessability, high dielectric strength and good cross-linking properties. It has turned out, though, that for various reasons existing low pressure materials are not suitable as replacement for LDPE as insulating material
5 for electric cables.

Thus, conventional high density polyethylene (HDPE) produced by polymerisation with a coordination catalyst of Ziegler-Natta type at low pressure has a melting point of about 130-135°C. When a HDPE is processed in an extruder the temperature should lie above the melting point of
10 130-135°C to achieve good processing. This temperature lies above the decomposition temperature of the peroxides used for the crosslinking of insulating ethylene polymer compositions. Dicumyl peroxide e.g. which is the most
15 frequently used crosslinking peroxide starts to decompose at a temperature of about 135°C. Therefore, when HDPE is processed above its melting temperature in an extruder the crosslinking peroxide decomposes and prematurely crosslinks the polymer composition, a phenomenon referred
20 to as "scorching". If, on the other hand the temperature is kept below the decomposition temperature of the peroxide then the HDPE will not melt adequately and unsatisfactorily processing will result.

Further, ethylene copolymers made by polymerisation
25 with a coordination catalyst at low pressure, like linear low density polyethylene (LLDPE) are unsuitable due to poor processability. The processability may be improved by polymerising the LLDPE in two or more steps (bimodal or multimodal LLDPE), but such LLDPE includes high melting
30 HDPE fractions or components, particularly when the polymerisation is carried out with conventional Ziegler-Natta catalysts, which makes LLDPE unsuitable for the same reason as conventional HDPE.

In this connection WO 93/04486 discloses an electrically
35 rically conductive device having an electrically conductive member comprising at least one electrically insulating member. The insulating member comprises an

ethylene copolymer with a density of 0.86-0.96 g/cm³, a melt index of 0.2-100 dg/min, a molecular weight distribution of 1.5-30, and a composition distribution breadth index (CDBI) greater than 45%. The copolymer of this
5 reference is unimodal as opposed to multimodal.

WO 97/50093 discloses a tree resistant cable comprising an insulation layer further comprising a multimodal copolymer of ethylene, said copolymer having a broad comonomer distribution as measured by TREF, a low
10 WTGR value and specified MFR and density values. Moreover, a low dissipation factor is disclosed. The document does not discuss the problem of premature decomposition of the crosslinking peroxide.

EP-A-743161 discloses a process for coextruding an
15 insulation layer and a jacketing layer on a conductive medium. The insulation layer is a metallocene based polyethylene having a narrow molecular weight distribution and a narrow comonomer distribution. The document further reveals that the extrusion of the narrow molecular weight
20 polymer at a low temperature is likely to lead to melt flow irregularities (so called melt fracture). This problem can be overcome by coextruding the insulation and the jacketing layer simultaneously on the conductor.

WO 98/41995 discloses a cable where the conductor is
25 surrounded by an insulation layer comprising a mixture of a metallocene based PE having a narrow molecular weight distribution and a narrow comonomer distribution and a low density PE produced in a high pressure process. The
addition of LDPE in metallocene PE is necessary to avoid
30 the melt flow irregularities, which are the result of the narrow molecular weight distribution of the metallocene PE.

In view of the above it would be an advantage if it was possible to replace crosslinkable LDPE made by
35 radical initiated polymerisation as a material for the insulating layer of electric power cables by an ethylene polymer made by coordination catalysed low pressure poly-

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merisation. Such a replacement polymer should have rheological properties, including processability similar to those of LDPE. Further, it should have a low enough melting temperature to be completely melted at 125°C in order to avoid "scorch" due to premature decomposition of the crosslinking peroxide.

Summary of the invention

It has now been discovered that LDPE may be replaced as a crosslinkable material for the insulation layer of electric cables by a crosslinkable ethylene copolymer made by coordination catalysed low pressure polymerisation which ethylene copolymer is a multimodal ethylene copolymer with specified density and viscosity and with melting temperature of at most 125°C.

More particularly the present invention provides an insulating composition for an electric power cable which comprises a crosslinkable ethylene polymer, characterised in that the ethylene polymer is a multimodal ethylene copolymer obtained by coordination catalysed polymerisation of ethylene and at least one other alpha-olefin in at least one stage, said multimodal ethylene copolymer having a density of 0.890-0.940 g/cm³, a MFR₂ of 0.1-10 g/10 min a MWD of 3.5-8, a melting temperature of at most 125°C and a comonomer distribution as measured by TREF, such that the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 10% by weight, and said multimodal ethylene copolymer including an ethylene copolymer fraction selected from (a) a low molecular weight ethylene copolymer having a density of 0.900-0.950 g/cm³ and a MFR₂ of 25-500 g/10 min, and (b) a high molecular weight ethylene copolymer having a density of 0.870-0.940 g/cm³ and a MFR₂ of 0.01-3 g/10 min.

Preferably, the polymer has a viscosity of 2500-7500 Pa.s at 135°C and a shear rate of 10 s⁻¹ 1000-2200 Pa.s at 135°C and a shear rate of 100 s⁻¹ 250-400 Pa.s at 135°C and a shear rate of 1000 s⁻¹.

A density in the lower part of the range, i.e. 0.890-0.910 g/cm³ is aimed at when a very flexible cable is desired. Such cables are suitable for applications in cars, mines and the building industry. These low densities are only possible to reach by using a single site catalyst such as a metallocene type catalyst, at least for the higher molecular weight fraction. When densities in the range 0.910-0.940 g/cm³ are chosen, the resulting cables are stiffer, but have better mechanical strength values, and are therefore more suitable for non-flexible power supply cables.

The present invention also provides an electric power cable comprising a conductor surrounded by an inner semiconducting layer, an insulating layer, and an outer semiconducting layer, characterised in that the insulating layer comprises a crosslinked ethylene copolymer obtained by coordination catalysed polymerisation of ethylene and at least one other alpha-olefin in at least one stage, said multimodal ethylene copolymer having a density of 0.890-0.940 g/cm³, a MFR₂ of 0.1-10 g/10 min, a MWD of 3.5-8, a melting temperature of at most 125°C and a comonomer distribution as measured by TREF such that the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 10% by weight, and said multimodal ethylene copolymer including an ethylene copolymer fraction selected from (a) a low molecular weight ethylene copolymer having a density of 0.900-0.950 g/cm³ and a MFR₂ of 25-500 g/10 min, and (b) a high molecular weight ethylene copolymer having a density of 0.870-0.940 g/cm³ and a MFR₂ of 0.01-3 g/10 min.

Preferably, the polymer has a viscosity of 2500-7500 Pa.s at 135°C and a shear rate of 10 s⁻¹ 1000-2200 Pa.s. at 135°C and a shear rate of 100 s⁻¹ and 250-400 Pa.s at 135°C and a shear rate of 1000 s⁻¹.

These and other characteristics of the invention will appear from the appended claims and the following description.

Detailed description of the invention

Before the invention is described in more detail, some key expressions will be defined.

By the "modality" of a polymer is meant the structure of the molecular-weight distribution of the polymer, i.e. the appearance of the curve indicating the number of molecules as a function of the molecular weight. If the curve exhibits one maximum, the polymer is referred to as "unimodal", whereas if the curve exhibits a very broad maximum or two or more maxima and the polymer consists of two or more fractions, the polymer is referred to as "bimodal", "multimodal" etc. In the following, all polymers which consist of at least two fractions and the molecular-weight-distribution curves of which are very broad or have more than one maximum are jointly referred to as "multimodal".

By the expression "melt flow rate" (MFR) used herein is meant, unless otherwise stated, the melt flow rate of a polymer as determined in accordance with ISO 1133, condition 4 (MFR₂). The melt flow rate, which is indicated in g/10 min, is an indication of the flowability, and hence the processability, of the polymer. The higher the melt flow rate, the lower the viscosity of the polymer.

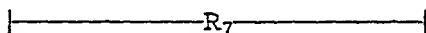
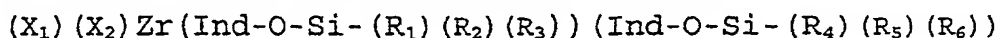
The expression "coordination catalyst" encompasses catalysts of the Ziegler-Natta type and single site catalysts, such as metallocene catalysts.

The "molecular weight distribution" (MWD) of a polymer means its molecular weight distribution as determined by the ratio between the weight average molecular weight (M_w) and the number average molecular weight (M_n) of the polymer (M_w/M_n).

It is previously known to produce multimodal, in particular bimodal, olefin polymers, preferably multimodal ethylene plastics, in two or more reactors connected in series. As instances of this prior art, mention may be made of EP 040 992, EP 041 796, EP 022 376 and WO 92/12182, which are hereby incorporated by way of reference as

regards the production of multimodal polymers. According to these references, each and every one of the polymerisation stages can be carried out in liquid phase, slurry or gas phase.

- 5 The catalyst used to produce the composition is a supported single site catalyst. The catalyst should produce a relatively narrow molecular weight distribution and comonomer distribution in one stage polymerisation. Also, the catalyst should be able to produce a high
 10 enough molecular weight so that good mechanical properties are obtained. It is known that some metallocene catalysts are able to produce a high enough molecular weight. Examples of such catalysts are e.g. those based on siloxy-substituted bridged bis-indenyl zirconium
 15 dihalides, as disclosed in the Finnish patent application FI 960437 which have the general formula:



where

- 20 X_1 and X_2 are either same or different and are selected from a group containing halogen, methyl, benzyl and hydrogen,
 Zr is zirconium,
 Ind is an indenyl group,
 25 R_1 to R_6 are either the same or different and are selected from a group containing linear and branched hydrocarbyl groups containing 1-10 carbon atoms and hydrogen,
 R_7 is a linear hydrocarbyl group containing 1-10 carbon atoms,
 30 Si is silicon, and
 O is oxygen;
 or on n-butyl dicyclopentadienyl hafnium compounds disclosed in FI-A-934917 which have the general formula:
 $(X_1)(X_2)Hf(Cp-R_1)(Cp-R_2)$
 35 where

X_1 and X_2 are either same or different and are selected from a group containing halogen, methyl, benzyl or hydrogen,

Hf is hafnium

5 Cp is cyclopentadienyl group, and

R_1 and R_2 are either the same or different and are either linear or branched hydrocarbyl groups containing 1-10 carbon atoms.

These catalysts may be supported on any known support material, such as silica, alumina, silica-alumina etc. Preferably, the catalyst is supported on silica. They are used together with an aluminumoxane cocatalyst. Examples of these cocatalysts are e.g. methylaluminumoxane (MAO), tetraisobutylaluminumoxane (TIBAO) and
15 hexaisobutylaluminumoxane (HIBAO). The cocatalyst is supported on the carrier, preferably together with the catalyst complex.

When the aluminumoxane cocatalyst is supported on the carrier with the metallocene complex, a lower amount
20 of the cocatalyst is needed than when it is introduced into the reactor separately. This is especially advantageous for a cable insulation material, since the low metal content results in a low dissipation factor. At the present invention the total metal contents (such as Al+Zr
25 or Al+Hf) in the polymer preferably is less than 70 ppm, more preferably less than 50 ppm.

According to the present invention, the main polymerisation stages are preferably carried out as a combination of slurry polymerisation/gas-phase polymerisation or gas-phase polymerisation/gas-phase polymerisation. The slurry polymerisation is preferably performed
30 in a so-called loop reactor. The use of slurry polymerisation in a stirred-tank reactor is not preferred in the present invention, since such a method is not sufficiently flexible for the production of the inventive composition and involves solubility problems. In order to produce the inventive composition, a flexible method is
35

required. For this reason, it is preferred that the composition is produced in two main polymerisation stages in a combination of loop reactor/gas-phase reactor or gas-phase reactor/gas-phase reactor. It is especially preferred that the composition is produced in two main polymerisation stages, in which case the first stage is performed as slurry polymerisation in a loop reactor and the second stage is performed as gas-phase polymerisation in a gas-phase reactor. Optionally, the main polymerisation stages may be preceded by a prepolymerisation, in which case up to 20% by weight, preferably 1-10% by weight, of the total amount of polymers is produced. Generally, this technique results in a multimodal polymer through polymerisation with the aid of a single site catalyst such as a metallocene catalyst in several successive polymerisation reactors.

Alternatively, a multimodal polymer may be produced through polymerisation in one single polymerisation reactor with the aid of a dual site coordination catalyst or a blend of different coordination catalysts. The dual site catalyst may comprise two or more different single site or metallocene species, each one of which produces a narrow molecular weight distribution and a narrow comonomer distribution. If a blend of catalysts is used, they need to be of a single site type of catalysts, such as metallocene catalysts. It is preferred, though, that the polymerisation be carried out in two or more polymerisation reactors connected in series.

In the production of a bimodal ethylene copolymer, a first ethylene copolymer fraction is produced in a first reactor under certain conditions with respect to monomer composition, hydrogen-gas pressure, temperature, pressure, and so forth. After the polymerisation in the first reactor, the reaction mixture including the copolymer fraction produced is fed to a second reactor, where further polymerisation takes place under other conditions. Usually, a first copolymer fraction of high melt flow rate (low

molecular weight) and with an addition of comonomer, is produced in the first reactor, whereas a second copolymer fraction of low melt flow rate (high molecular weight) and with an addition of comonomer is produced in the second
5 reactor. As comonomer, use is preferably made of α -olefins having up to 8 carbon atoms, such as propene, 1-butene, 4-methyl-1-pentene, 1-hexene, and 1-octene. The resulting end product consists of an intimate mixture of the copolymers from the two reactors, the different molecular-
10 -weight-distribution curves of these copolymers together forming a molecular-weight-distribution curve having one broad maximum or two maxima, i.e. the end product is a bimodal polymer mixture. Since multimodal, and especially bimodal polymers, and the production thereof belong to the
15 prior art, no further detailed description is called for here, but reference is made to the above specifications.

It should here be pointed out that, in the production of two or more polymer components in a corresponding number of reactors connected in series, it is only in the
20 case of the component produced in the first reactor stage and in the case of the end product that the melt flow rate, the density and the other properties can be measured directly on the material removed. The corresponding properties of the polymer components produced in
25 reactor stages following the first stage can only be indirectly determined on the basis of the corresponding values of the materials introduced into and discharged from the respective reactor stages.

Even though multimodal polymers and their production
30 are known per se, it is not previously known to prepare multimodal copolymers having the specific characteristics defined above and use them as insulating layers for electric power cables.

As hinted above, it is preferred that the multimodal
35 olefin copolymer in the cable-insulating composition according to the invention is a bimodal ethylene copolymer. It is also preferred that this bimodal ethylene

copolymer has been produced by polymerisation as above under different polymerisation conditions in two or more polymerisation reactors connected in series. Owing to the flexibility with respect to reaction conditions thus obtained, it is preferred that the polymerisation is carried out in a loop reactor/a gas-phase reactor, a gas-phase reactor/a gas-phase reactor or a loop reactor/a loop reactor. The polymerisation conditions in the preferred two-stage method are so chosen that a comparatively low molecular weight ethylene copolymer is produced in one stage, preferably the first stage, owing to a high content of chain-transfer agent (hydrogen gas), whereas a high molecular weight ethylene copolymer is produced in another stage, preferably the second stage. The order of these stages may, however, be reversed.

As mentioned above, the multimodal ethylene copolymer of the invention should have a density of 0.890-0.940 g/cm³.

Further, the comonomer content of the multimodal ethylene copolymer of the invention should lie within the range 2-22 % by weight based on the copolymer. As the density of the copolymer is related to the comonomer content and is roughly inversely proportional to the comonomer content, this means that the lower density of 0.890 g/cm³ corresponds to the higher comonomer content of about 18% by weight, whereas the higher density corresponds to the lower comonomer content of 2% by weight.

As stated earlier, the comonomer of the ethylene copolymer of the present invention is selected from other alpha-olefins, preferably other C₃-C₈ alpha-olefins. It is particularly preferred that the comonomer is selected from at least one member of the group consisting of propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, and 1-octene.

The comonomer distribution of the polymer composition should be such that the composition does not contain high density polyethylene having a high melting temperature. This is the case if, when the composition is ana-

lyzed by TREF, the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 10%. Preferably, the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 7% and in particular, no more than 5% of the copolymer elutes at a temperature higher than 90°C.

As is seen from the enclosed TREF fractograms of Figs 1 and 2 of Examples 3 and 4, respectively, the TREF fractogram of the copolymer according to the invention preferably contains two separate peaks.

It is an essential characteristic of the multimodal ethylene copolymer of the present invention that it has a melting temperature (T_m) of at most 125°C. This means that the multimodal ethylene copolymer does not contain any ethylene copolymer fraction with a melting temperature above 125°C.

Another essential characteristic of the multimodal ethylene copolymer of the present invention is that its processing properties are similar to those of LDPE. More particularly, the multimodal ethylene copolymer of the invention preferably has a viscosity of

2500-7500 Pa.s at 135°C and a shear rate of 10 s⁻¹,
1000-2200 Pa.s at 135°C and a shear rate of 100 s⁻¹,

and

250-400 Pa.s at 135°C and a shear rate of 1000 s⁻¹.

More preferably, the viscosity is as follows:

4000-7000 Pa.s at 135°C and a shear rate of 10 s⁻¹,
1000-2000 Pa.s at 135°C and a shear rate of 100 s⁻¹,

and

300-350 Pa.s at 135°C and a shear rate of 1000 s⁻¹.

The above viscosity values illustrate the processing behaviour of the multimodal ethylene copolymer of the invention very well. Further, the viscosity of the multimodal ethylene copolymer determined by its melt flow rate, MFR₂, should lie in the range 0.1-10.0, preferably 0.5-7.0 g/10 min, more preferably 0.5-3.0 g/10 min, and most preferably 1.0-3.0 g/10 min.

The multimodal ethylene copolymer of the invention has a molecular weight distribution, MWD, of 3.5-8, preferably 3.5-6, more preferably 4-6, and in particular 4-5.

5 In order to be crosslinkable the multimodal ethylene copolymer of the present invention should have a degree of unsaturation of at least about 0.3-0.6 double bonds/1000 carbon atoms.

10 The multimodal ethylene copolymer is made up of at least two ethylene copolymer fractions and the properties of the individual copolymer fractions should be so chosen that the above specified values of density/comonomer content, viscosity/melt flow rate, MWD and melting temperature of the multimodal ethylene copolymer are achieved.

15 Although the multimodal ethylene copolymer of the invention could in principle consist of a polymerised blend of any number of ethylene copolymer fractions, it is preferred that it consists of two ethylene copolymer fractions only, namely a low molecular weight (LMW)
20 ethylene copolymer fraction and a high(er) molecular weight (HMW) ethylene copolymer fraction.

The preferred multimodal ethylene copolymer of the invention is thus obtained by a two-stage polymerisation process, where a LMW ethylene copolymer fraction is
25 produced in the first polymerisation stage and a HMW ethylene copolymer fraction is produced in the second polymerisation stage. Preferably for use in non-flexible power supply cable, the LMW ethylene copolymer fraction has a density of 0.925-0.940 g/cm³, and a MFR₂ of 25-300,
30 preferably 40-200, more preferably 50-100 g/10 min. For use in flexible applications the density should preferably lie in the range 0.900-0.925 g/cm³. The comonomer content of the LMW ethylene copolymer fraction is preferably 3-15 % by weight. The HMW ethylene copolymer
35 fraction has such a density, comonomer content, and MFR that the multimodal ethylene copolymer obtains the values

of density/comonomer content, viscosity/melt flow rate, MWD and melting temperature specified above.

For use in flexible cable, it is preferred that the LMW fraction has a lower density of 0.900-0.925 g/cm³ but similar MFR₂-values as for non-flexible cable applications.

More particularly, a calculation indicates that when the LMW ethylene copolymer has the above specified values, the HMW ethylene copolymer produced in the second polymerisation stage of a two-stage process should have a density of 0.870-0.910 g/cm³ for flexible cable and of 0.910-0.940 g/cm³ for non-flexible cable, and a MFR₂ of 0.01-3, preferably 0.1-2.0 g/10 min. Preferably the comonomer content is 20-15 % by weight in flexible compositions and 18-2 % by weight in non-flexible ones.

As stated in the foregoing, the order of the polymerisation stages may be reversed, which would mean that, if the multimodal ethylene copolymer has a density and a viscosity as defined above, and the HMW ethylene copolymer produced in the first polymerisation stage has a density of 0.910-0.940 g/cm³ for non-flexible applications and 0.870-0.910 g/cm³ for flexible ones, and a MFR₂ of 0.01-3 g/10 min, then the LMW ethylene copolymer produced in the second polymerisation stage of a two-stage process should, according to calculations as above, have a density of 0.920-0.950 g/cm³ for non-flexible compositions and of 0.900-0.930 g/cm³ for flexible ones, and a MFR₂ of 25-300 g/10 min. This order of the stages in the production of the multimodal ethylene copolymer according to the invention is, however, less preferred.

In the multimodal ethylene copolymer of the invention the LMW ethylene copolymer fraction preferably comprises 30-60 % by weight of the multimodal ethylene copolymer and, correspondingly, the HMW ethylene copolymer fraction comprises 70-40 % by weight.

Besides the multimodal ethylene copolymer and a cross-linking agent the insulating composition of the present invention may include various additives commonly employed

In order to further illustrate the present invention
5 and facilitate its understanding some non-restricting
Examples are given below.

MFR₂ determined at 190°C using 2.16 kg load, according to ISO 1133.

TREF (Temperature Rising Elution Fractionation) described in L. Wild, T.R. Ryle, D.C. Knobeloch and I.R. Peak in Journal of Polymer Science: Polymer Physics Edition, Vol. 20, 441-455 (1982).

Contents of Al, Zr and Hf were determined by AAS (Atomic Adsorption Spectroscopy).

Example 1

The thus obtained catalyst was dried under nitrogen for 6 hours at 75°C temperature. Then, the catalyst was further dried under vacuum for 10 hours.

168 g of a metallocene complex (ethylene bridged
siloxo-substituted bis-indenyl zirconium dichloride accord-

ing to the patent application FI 960437) and 9.67 kg of a 30% MAO solution supplied by Albemarle were combined and 3.18 kg dry purified toluene was added. The thus obtained complex solution was added on 9 kg silica carrier Sylopol 55 SJ by Grace. The complex was fed very slowly with uniform spraying during 2 hours. Temperature was kept below 30°C. The mixture was allowed to react for 2 hours after complex addition at 30°C.

The thus obtained catalyst was dried under nitrogen for 6 hours at 75°C temperature. Then, the catalyst was further dried under vacuum for 10 hours.

Example 3

Into a loop reactor having a volume of 500 dm³ was introduced a polymerization catalyst prepared according to Example 1, propane diluent, ethylene, 1-butene comonomer and hydrogen. The reactor was operated at 85°C temperature and 60 bar pressure. The feed rates of the components were such that 25 kg/h of polyethylene having MFR₂ of 85 g/10 min and density 934 kg/m³ was formed. The polymer containing the active catalyst was separated from the reaction media and transferred to a gas phase reactor operated at 75°C temperature and 20 bar pressure, where additional ethylene, hydrogen and 1-butene comonomer were added so, that in total 60 kg/h polyethylene having MFR₂ of 2.6 g/10 min and density 913 kg/m³ was collected from the reactor. The fraction of the high MFR material (or low molecular weight material) in the total polymer was thus 42%.

The metal contents of the polymer were analyzed. The total ash content was 390 ppm, the Hf content was 1 ppm and the Al content was 35 ppm.

The viscosity of the polymer was measured at 10, 100 and 1000 s⁻¹ shear rates. They were found to be 5600, 2000 and 360 Pa.s respectively.

The polymer was analyzed by using TREF. The analysis revealed that 4.8% of the polymer eluted at a temperature higher than 90°C and 1.2% eluted at a temperature higher than 95°C (cf. Fig. 1).

Dissipation factor of the material was measured from 3.0 mm thick compression moulded plaques at 500 V. It was found to be $2.0 \cdot 10^{-4}$ and $0.9 \cdot 10^{-4}$ as measured immediately after compression moulding and after 3 days aging, respectively.

Example 4

The polymerization was conducted as in Example 3, with the exception that a catalyst prepared according to Example 2 was used and that the temperature of the loop reactor was 75°C. In the loop reactor 25 kg/h of polyethylene having MFR₂ of 260 g/10 min and density 931 kg/m³ was formed. The polymer containing the active catalyst was separated from the reaction media and transferred to a gas phase reactor operated at 75°C temperature and 20 bar pressure, where additional ethylene, hydrogen and 1-butene comonomer were added so, that in total 52 kg/h polyethylene having MFR₂ of 1.4 g/10 min and density 918 kg/m³ was collected from the reactor. The fraction of the high MFR material in the total polymer was thus 48%.

The metal contents of the polymer were analyzed. The total ash content was 190 ppm, the Zr content was less than 1 ppm and the Al content was 15 ppm.

The viscosity of the polymer was measured at 10, 100 and 1000 s⁻¹ shear rates. They were found to be 6200, 1700 and 330 Pa.s respectively.

The polymer was analyzed by using TREF. The analysis revealed that 4.5% of the polymer eluted at a temperature higher than 90°C and 0.8% eluted at a temperature higher than 95°C (cf. Fig. 2).

Dissipation factor of the material was measured from 3.0 mm thick compression moulded plaques at 500 V. It was found to be $0.9 \cdot 10^{-4}$ and $0.4 \cdot 10^{-4}$ as measured immediately after compression moulding and after 3 days aging, respectively.

Example 5

The polymerization was conducted as in Example 4. In the loop reactor 25 kg/h of polyethylene having MFR₂ of

150 g/10 min and density 929 kg/m³ was formed. The polymer containing the active catalyst was separated from the reaction media and transferred to a gas phase reactor, where additional ethylene, hydrogen and 1-butene comonomer were added so, that in total 52 kg/h polyethylene having MFR₂ of 1.2 g/10 min and density 915 kg/m³ was collected from the reactor. The fraction of the high MFR material in the total polymer was thus 48%.

The metal contents of the polymer were analyzed. The total ash content was 190 ppm, the Zr content was less than 1 ppm and the Al content was 13 ppm.

The viscosity of the polymer was measured at 10, 100 and 1000 s⁻¹ shear rates. They were found to be 6800, 1800 and 360 Pa.s respectively.

The polymer was analyzed by using TREF. The analysis revealed that 4.2% of the polymer eluted at a temperature higher than 90°C and 0.7% eluted at a temperature higher than 95°C. Dissipation factor of the material was measured from 3.0 mm thick compression moulded plaques at 500 V. It was found to be $0.8 \cdot 10^{-4}$ and $0.5 \cdot 10^{-4}$ as measured immediately after compression moulding and after 3 days aging, respectively.

Example 6

Into a sample of material produced in Example 3 was added 0.2-% by weight of 4.4'-thio-bis-(2-tert-butyl-5-methyl-phenol) stabiliser and 1.9 wt-% dicumylperoxide (used as a crosslinking agent). The composition was then compounded at a melt temperature of about 130°C. The crosslinking properties of the insulating composition were evaluated by the hot set test. In this test the elongation of dumbbells was measured at 200°C with a load of 0.2 MPa. The elongation was found to be 37% and the permanent deformation was found to be 1%.

Example 7

The procedure of Example 6 was repeated, except that a material produced in Example 4 was used. It should be noted that the material of Example 4 contained 0.1% by weight of

Irganox B561 stabilizer. In the hot set test the elongation was 25% and the permanent deformation 0.3%. Hot set data is summarized in Table 1:

Table 1

	Elongation %	Permanent deformation %
Example 6	37	1
Example 7	25	0.3
Comparative Example 1	33	1

5 From the table it can be concluded that the degree of crosslinking in Examples 6 and 7 with peroxide content of 1.9% was equal to that Comparative Example 1 with peroxide content of 2.0%.

Example 8

10 A model cable was produced using the composition according to Example 6 as an insulation layer. The model cable was produced by using of a triple extruder head where an inner semiconductive layer, insulation layer and an
15 the conductor without difficulties. The semiconductive layers comprised a crosslinkable ethylene-butyl acrylate copolymer (17% by weight of BA) containing about 40% by weight of carbon black. The thickness: inner semicon layer was 0.7 mm, the thickness of the insulation layer was
20 1.5 mm and the thickness of the outer semicon layer was 0.15 mm.

The layers were coextruded through a triple head extruder onto a conductor using a temperature setting ranging from 105-130°C.

25 Example 9

The procedure of Example 8 repeated, except that a composition produced in Example 7 was used. The data of Examples 8 and 9 and Comparative Example 1 is shown in Table 2. Also the values of dissipation factor are shown
30 for Examples 8 and 9.

Table 2

	F _{min} Nm	T ₁₀ min	Tan δ
Example 8	36	69	$1.2 \cdot 10^{-4}$
Example 9	43	62	$1.0 \cdot 10^{-4}$
Comparative Example 1	81	26	

The results show that the material according to the present invention has a better scorch resistance (higher value of T₁₀) compared to a comparative material at an equal degree of crosslinking.

Comparative Example 1

For the polymerisation of ethylene a loop reactor and a gas-phase reactor connected in series were used together with a prepolymerisation reactor (Pre PR). In addition to ethylene 1-butene was used as a comonomer in the loop reactor and the gas-phase reactor. Hydrogen was used as a modifier. The catalyst was a catalyst of Ziegler-Natta type and was added to the prepolymerisation reactor. Propane was used as a reaction medium in the loop reactor. The gaseous components of the product from the loop reactor were removed in a flash tank, whereafter the product was transferred to the gas-phase reactor where the polymerisation was continued. The polymerisation conditions and the product properties are shown in Table 3.

The material was analyzed by using TREF. It revealed that 26.1% of the polymer eluted at a temperature above 90°C and 12.8% of the material eluted at a temperature above 95°C.

After compounding the copolymer with 0.2% by weight of Santonox R (a stabiliser), and adding 2.0 % by weight of dicumyl peroxide (crosslinking agent) the crosslinking properties of the insulating composition were evaluated by the hot set test. In the hot set test, the elongation of dumbbells was measured at 200°C with a load of 0.2 MPa. Decaline extraction was performed ASTM D 2765. The results are given in Table 4.

Table 4 also shows the results of scorch-testing. The measurements were performed on a Brabender Plasti-corder PL 2000-6 at 5 rpm at 135°C. The oil heated kneader 350, 287 cm³ with walzenkneaders W7646 was used.

- 5 The time to increase the torque value by 10 Nm (T_{10}) from the minimum value (F_{min}) was measured.

It is evident from Table 4 that the insulating composition according to this example which has too high viscosity and too high TREF values is somewhat scorch
10 sensitive. The T_{10} time of 26 min should be compared with about 56 min for conventional crosslinkable LDPE. The hot set elongation was good.

Table 3

<u>First reactor (PR1)</u>		
15	Temperature (°C)	85
	Pressure (MPa)	6.1
	Ethylene concentration (MPa)	0.66
	Hydrogen concentration (mol/kmol C ₂)	142
	1-butene concentration (mol/kmol C ₂)	630
20	Product density (g/cm ³)	0.943
	MFR ₂ (g/10 min)	230
<u>Second reactor (PR2)</u>		
	Temperature (°C)	75
	Pressure (MPa)	2.0
25	Ethylene concentration (MPa)	1.57
	Hydrogen concentration (mol/kmol C ₂)	30
	1-butene concentration (mol/kmol/C ₂)	500
	Split (product ratio PrePR:PR1:PR2)	1:42:57
<u>End product</u>		
30	Product density (g/cm ³)	0.926
	MFR ₂ (g/10 min)	0.53
	MWD	11.3
	Melting temperature (°C)	122
	Comonomer content (% by weight)	7.7
35	Degree of unsaturation (C=C/1000C)	0.27

Apparent viscosity (Pa.s)
at 135°C

	Shear rate: 10 s ⁻¹	7900
	Shear rate: 100 s ⁻¹	1900
5	Shear rate: 1000 s ⁻¹	360

Table 4

	Elongation/Set (%/%)	33/-1
	Gel content (%)	79.6
	Scorch T ₁₀ min	26
10	Scorch F _{min} (nm)	81

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AMENDED CLAIMS

1. An insulating composition for an electric power cable which comprises a crosslinkable ethylene polymer, characterised in that the ethylene polymer is a multimodal ethylene copolymer obtained by coordination
5 catalysed polymerisation of ethylene and at least one other alpha-olefin in at least one stage, said multimodal ethylene copolymer having a density of 0.890-0.940 g/cm³, a MFR₂ of 0.1-10 g/10 min, a MWD of 3.5-8, a melting temperature of at most 125°C, and a comonomer
10 distribution as measured by TREF, such that the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 5% by weight, and said multimodal ethylene copolymer including an ethylene copolymer fraction selected from (a) a low molecular weight
15 ethylene copolymer having a density of 0.900-0.950 g/cm³ and a MFR₂ of 25-500 g/10 min, and (b) a high molecular weight ethylene copolymer having a density of 0.870-0.940 g/cm³ and a MFR₂ of 0.01-3 g/10 min.

2. An insulating composition as claimed in claim 1,
20 wherein the multimodal ethylene copolymer has a comonomer distribution as measured by TREF such that the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 7% by weight.

3. An insulating composition as claimed in any one
25 of claims 1-2, wherein the multimodal ethylene copolymer has a viscosity of
2500-7500 Pa.s at 135°C and a shear rate of 10 s⁻¹,
1000-2200 Pa.s at 135°C and a shear rate of 100 s⁻¹, and
250-400 Pa.s at 135°C and a shear rate of 1000 s⁻¹.

30 4. An insulating composition as claimed in claim 3, wherein the multimodal ethylene copolymer has a viscosity of
4000-7000 Pa.s at 135°C and a shear rate of 10 s⁻¹,
1000-2000 Pa.s at 135°C and a shear rate of 100 s⁻¹, and
35 300-350 Pa.s at 135°C and a shear rate of 1000 s⁻¹.

10 -01- 2001

5. An insulating composition as claimed in any one of claims 1-4, wherein the comonomer of the copolymer is at least one member selected from the group consisting of propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, and 1-octene.

6. An insulating composition as claimed in any one of claims 1-5, wherein the MWD is 4-5.

7. An insulating composition as claimed in any one of claims 1-6, wherein the multimodal ethylene copolymer is a bimodal ethylene copolymer comprising 30-60 % by weight of a low molecular weight ethylene copolymer fraction and 70-40 % by weight of a high molecular weight ethylene copolymer fraction.

8. An insulating composition as claimed in any one of claims 1-7, wherein the multimodal ethylene copolymer includes a low molecular weight ethylene copolymer fraction having a density of 0.900-0.950 g/cm³ and a MFR₂ of 50-100 g/10 min.

9. An electric power cable comprising a conductor surrounded by an inner semiconducting layer, an insulating layer, and an outer semiconducting layer, characterised in that the insulating layer comprises a crosslinked ethylene copolymer obtained by coordination catalysed polymerisation of ethylene and at least one other alpha-olefin in at least one stage, said multimodal ethylene copolymer having a density of 0.890-0.940 g/cm³, a MFR₂ of 0.1-10 g/10 min, a MWD of 3.5-8, a melting temperature of at most 125°C, and a comonomer distribution as measured by TREF, such that the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 5% by weight, and said multimodal ethylene copolymer including an ethylene copolymer fraction selected from (a) a low molecular weight ethylene copolymer having a density of 0.900-0.950 g/cm³ and a MFR₂ of 25-500 g/10 min, and (b) a high molecular weight ethylene copolymer having a density of 0.870-0.940 g/cm³ and a MFR₂ of 0.01-3 g/10 min.

ABSTRACT

An insulating composition for an electric power cable, and an electric power cable comprising a conductor surrounded by an inner semiconducting layer, an insulating layer, and an outer semiconducting layer, where the insulating layer consists of said insulating composition, are disclosed. The insulating composition is characterised in that the ethylene polymer is a multimodal ethylene copolymer obtained by coordination catalysed polymerisation of ethylene and at least one other alpha-olefin in at least one stage, said multimodal ethylene copolymer having a density of 0.890-0.940 g/cm³, a MFR₂ of 0.1-10 g/10 min, a MWD of 3.5-8, a melting temperature of at most 125°C, and a comonomer distribution as measured by TREF, such that the fraction of copolymer eluted at a temperature higher than 90°C does not exceed 10% by weight, and said multimodal ethylene copolymer including an ethylene copolymer fraction selected from (a) a low molecular weight ethylene copolymer having a density of 0.900-0.950 g/cm³ and a MFR₂ of 25-500 g/10 min, and (b) a high molecular weight ethylene copolymer having a density of 0.870-0.940 g/cm³ and a MFR₂ of 0.01-3 g/10 min.

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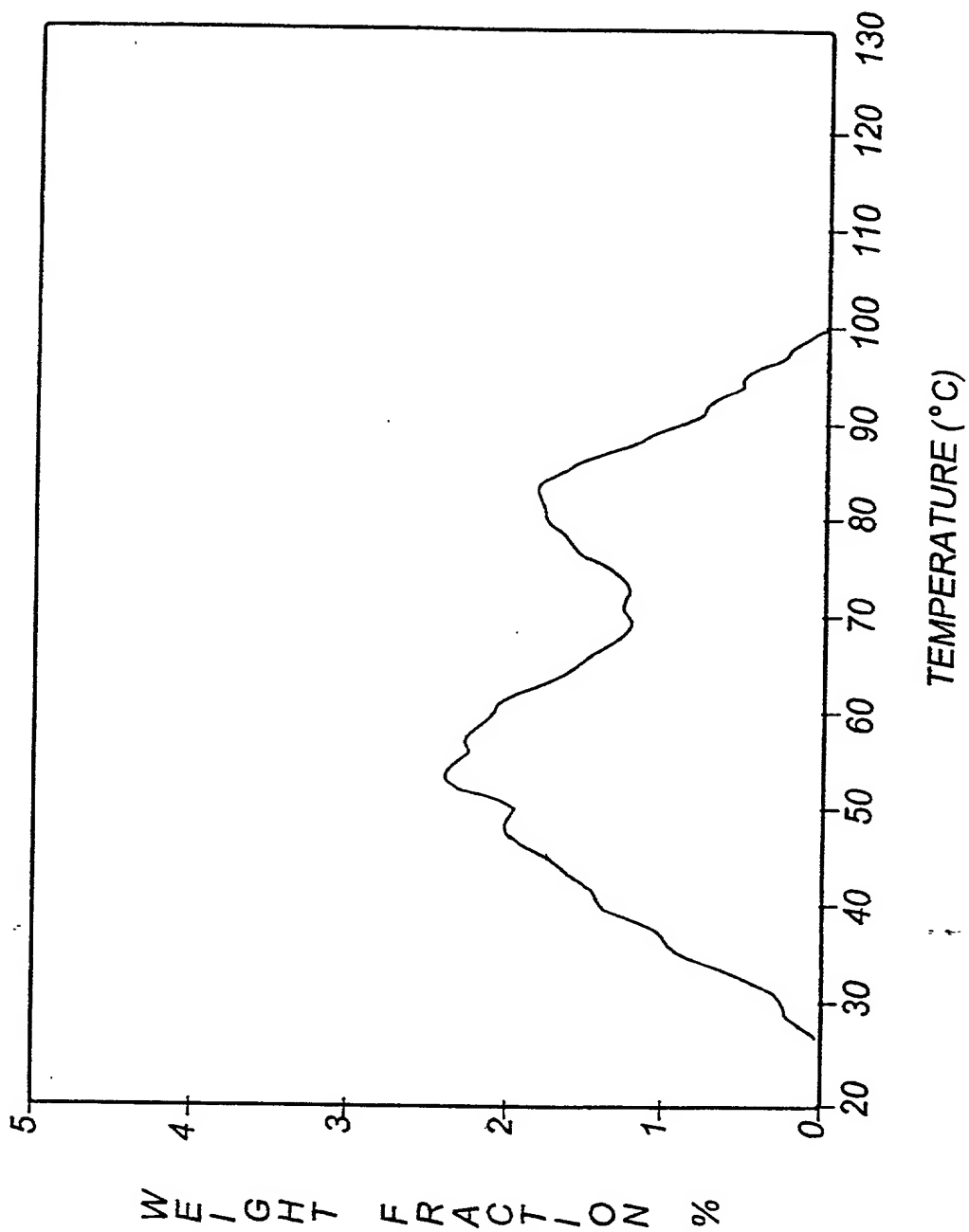


Fig. 1

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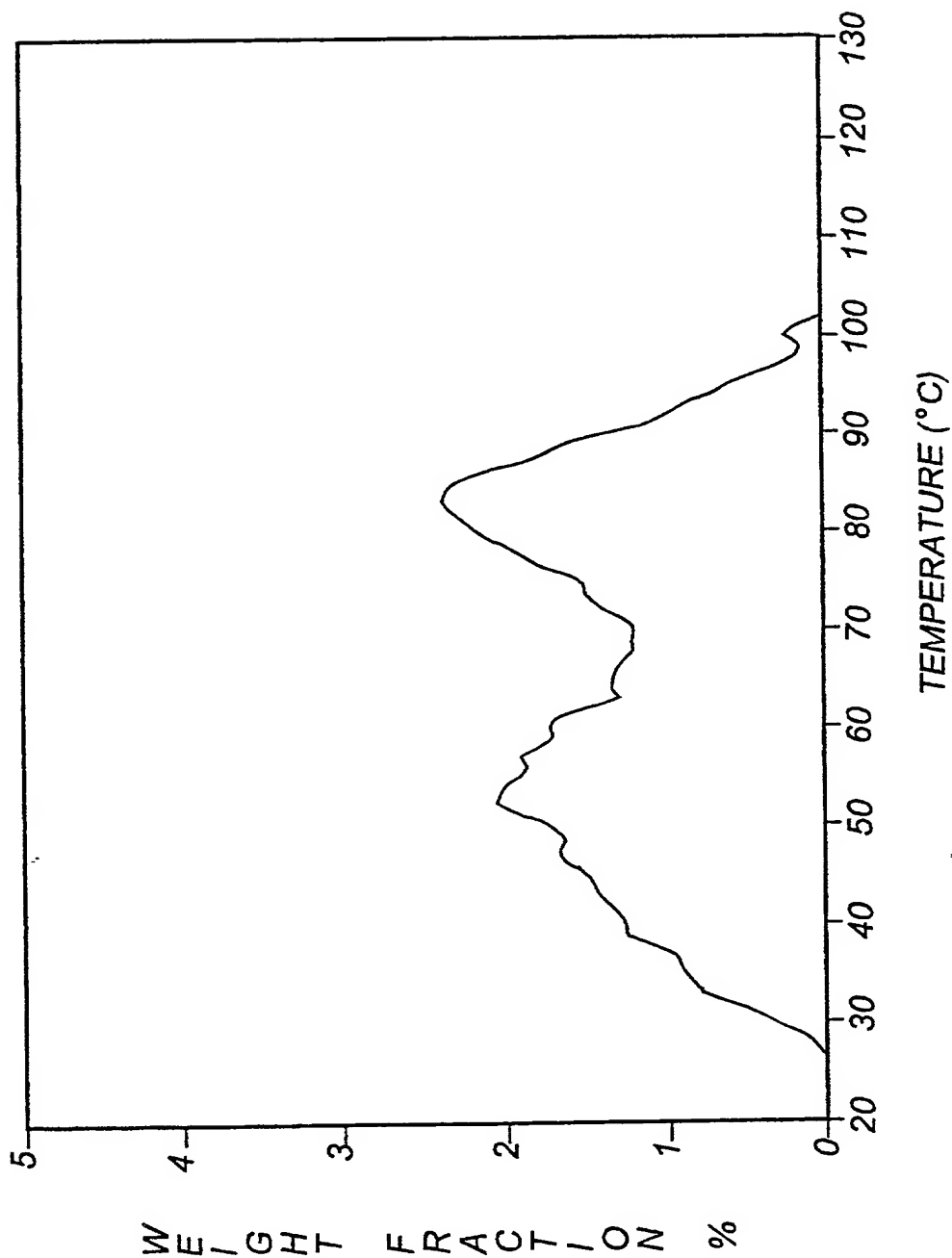


Fig. 2

F19939/US

Attorney Docket No. 5848.170USWO

MERCHANT & GOULD P.C.

United States Patent Application

COMBINED DECLARATION AND POWER OF ATTORNEY

As a below named inventor I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that

I verily believe I am the original, first and sole inventor (if only one name is listed below) or a joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled: AN INSULATION COMPOSITION FOR AN ELECTRIC CABLE

The specification of which

- a. ☐ is attached hereto
- b. ☒ was filed on 18 December 2001, as application serial no. _____ and was amended on _____ (if applicable) (in the case of a PCT-filed application) described and claimed in international no. PCT/SE00/01334 filed 22 June 2000, and as amended on 10 January 2001 (if any), which I have reviewed and for which I solicit a United States patent.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I hereby claim foreign priority benefits under Title 35, United States Code, § 119/365 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on the basis of which priority is claimed:

- a. ☐ no such applications have been filed.
- b. ☒ such applications have been filed as follows:

FOREIGN APPLICATION(S), IF ANY, CLAIMING PRIORITY UNDER 35 USC § 119			
COUNTRY	APPLICATION NUMBER	DATE OF FILING (day, month, year)	DATE OF ISSUE (day, month, year)
Sweden	9902510-8	01 July 1999	
ALL FOREIGN APPLICATION(S), IF ANY, FILED BEFORE THE PRIORITY APPLICATION(S)			
COUNTRY	APPLICATION NUMBER	DATE OF FILING (day, month, year)	DATE OF ISSUE (day, month, year)

I hereby claim the benefit under Title 35, United States Code, § 120/365 of any United States and PCT international application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, § 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. APPLICATION NUMBER	DATE OF FILING (day, month, year)	STATUS (patented, pending, abandoned)

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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 Glance, Robert J. Reg. No. 40,620
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 Goggin, Matthew J. Reg. No. 44,125
 Golla, Charles E. Reg. No. 26,896
 Gorman, Alan G. Reg. No. 38,472
 Gould, John D. Reg. No. 18,223
 Gregson, Richard Reg. No. 41,804
 Gressens, John J. Reg. No. 33,112
 Hamer, Samuel A. Reg. No. 46,754
 Hamre, Curtis B. Reg. No. 29,165
 Harrison, Kevin C. Reg. No. 46,759
 Hertzberg, Brett A. Reg. No. 42,660
 Hillson, Randall A. Reg. No. 31,838
 Holzer, Jr., Richard J. Reg. No. 42,668
 Hope, Leonard J. Reg. No. 44,774
 Jardine, John S. Reg. No. P-48,835
 Johns, Nicholas P. Reg. No. 48,995
 Johnston, Scott W. Reg. No. 39,721
 Kadievitch, Natalie D. Reg. No. 34,196
 Kaseburg, Frederick A. Reg. No. 47,695
 Kettelberger, Denise Reg. No. 33,924
 Keys, Jerramie J. Reg. No. 42,724
 Knearl, Homer L. Reg. No. 21,197
 Kowalchuk, Alan W. Reg. No. 31,535
 Kowalchuk, Katherine M. Reg. No. 36,848
 Lacy, Paul E. Reg. No. 38,946
 Larson, James A. Reg. No. 40,443
 Leonard, Christopher J. Reg. No. 41,940
 Liepa, Mara E. Reg. No. 40,066

Lindquist, Timothy A. Reg. No. 40,701
 Lown, Jean A. Reg. No. 48,428
 Mayfield, Denise L. Reg. No. 33,732
 McDonald, Daniel W. Reg. No. 32,044
 McIntyre, Jr., William F. Reg. No. 44,921
 Mitchem, M. Todd Reg. No. 40,731
 Mueller, Douglas P. Reg. No. 30,300
 Nelson, Anna M. Reg. No. 48,935
 Paley, Kenneth B. Reg. No. 38,989
 Parsons, Nancy J. Reg. No. 40,364
 Pauly, Daniel M. Reg. No. 40,123
 Phillips, John B. Reg. No. 37,206
 Pino, Mark J. Reg. No. 43,858
 Prendergast, Paul Reg. No. 46,068
 Pytel, Melissa J. Reg. No. 41,512
 Qualey, Terry Reg. No. 25,148
 Reich, John C. Reg. No. 37,703
 Reiland, Earl D. Reg. No. 25,767
 Samuels, Lisa A. Reg. No. 43,080
 Schmalz, David G. Reg. No. 39,828
 Schuman, Mark D. Reg. No. 31,197
 Schumann, Michael D. Reg. No. 30,422
 Scull, Timothy B. Reg. No. 42,137
 Sebald, Gregory A. Reg. No. 33,280
 Skoog, Mark T. Reg. No. 40,178
 Spellman, Steven J. Reg. No. 45,124
 Stewart, Alan R. Reg. No. 47,974
 Stoll-DeBell, Kirstin L. Reg. No. 43,164
 Sullivan, Timothy Reg. No. 47,981
 Summer, John P. Reg. No. 29,114
 Swenson, Erik G. Reg. No. 45,147
 Tellekson, David K. Reg. No. 32,314
 Trembath, Jon R. Reg. No. 38,344
 Tunheim, Marcia A. Reg. No. 42,189
 Underhill, Albert L. Reg. No. 27,403
 Vandenburg, J. Derek Reg. No. 32,179
 Wahl, John R. Reg. No. 33,044
 Weaver, Paul L. Reg. No. 48,640
 Welter, Paul A. Reg. No. 20,890
 Whipples, Brian Reg. No. 43,261
 Whitaker, John E. Reg. No. 42,222
 Wier, David D. Reg. No. P-48,229
 Williams, Douglas J. Reg. No. 27,054
 Withers, James D. Reg. No. 40,376
 Witt, Jonelle Reg. No. 41,980
 Wong, Thomas S. Reg. No. 48,577
 Wu, Tong Reg. No. 43,361
 Young, Thomas Reg. No. 25,796
 Zeuli, Anthony R. Reg. No. 45,255

I hereby authorize them to act and rely on instructions from and communicate directly with the person/assignee/attorney/firm/ organization who/which first sends/sent this case to them and by whom/which I hereby declare that I have consented after full disclosure to be represented unless/until I instruct Merchant & Gould P.C. to the contrary. I understand that the execution of this document, and the grant of a power of attorney, does not in itself establish an attorney-client relationship between the undersigned and the law firm Merchant & Gould P.C., or any of its attorneys.

I acknowledge the duty to disclose information that is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, § 1.56 (reprinted below):

§ 1.56 Duty to disclose information material to patentability.

(a) A patent by its very nature is affected with a public interest. The public interest is best served, and the most effective patent examination occurs when, at the time an application is being examined, the Office is aware of and evaluates the teachings of all information material to patentability. Each individual associated with the filing and prosecution of a patent application has a duty of candor and good faith in dealing with the Office, which includes a duty to disclose to the Office all information known to that individual to be material to patentability as defined in this section. The duty to disclose information exists with respect to each pending claim until the claim is canceled or withdrawn from consideration, or the application becomes abandoned. Information material to the patentability of a claim that is canceled or withdrawn from consideration need not be submitted if the information is not material to the patentability of any claim remaining under consideration in the application. There is no duty to submit information which is not material to the patentability of any existing claim. The duty to disclose all information known to be material to patentability is deemed to be satisfied if all information known to be material to patentability of any claim issued in a patent was cited by the Office or submitted to the Office in the manner prescribed by §§ 1.97(b)-(d) and 1.98. However, no patent will be granted on an application in connection with which fraud on the Office was practiced or attempted or the duty of disclosure was violated through bad faith or intentional misconduct. The Office encourages applicants to carefully examine:

(1) prior art cited in search reports of a foreign patent office in a counterpart application, and

(2) the closest information over which individuals associated with the filing or prosecution of a patent application believe any pending claim patentably defines, to make sure that any material information contained therein is disclosed to the Office.

(b) Under this section, information is material to patentability when it is not cumulative to information already of record or being made of record in the application, and

(1) It establishes, by itself or in combination with other information, a prima facie case of unpatentability of a claim;

(2) It refutes, or is inconsistent with, a position the applicant takes in:

(i) Opposing an argument of unpatentability relied on by the Office, or

(ii) Asserting an argument of patentability.

A prima facie case of unpatentability is established when the information compels a conclusion that a claim is unpatentable under the preponderance of evidence, burden-of-proof standard, giving each term in the claim its broadest reasonable construction consistent with the specification, and before any consideration is given to evidence which may be submitted in an attempt to establish a contrary conclusion of patentability.

(c) Individuals associated with the filing or prosecution of a patent application within the meaning of this section are:

(1) Each inventor named in the application:

(2) Each attorney or agent who prepares or prosecutes the application; and

(3) Every other person who is substantively involved in the preparation or prosecution of the application and who is associated with the inventor, with the assignee or with anyone to whom there is an obligation to assign the application.

(d) Individuals other than the attorney, agent or inventor may comply with this section by disclosing information to the attorney, agent, or inventor.

(e) In any continuation-in-part application, the duty under this section includes the duty to disclose to the Office all information known to the person to be material to patentability, as defined in paragraph (b) of this section, which became available between the filing date of the prior application and the national or PCT international filing date of the continuation-in-part application.